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#### FOREWARD

This report was prepared by the Space and Defense Sciences Department, Research Division, Allis-Chalmers Manufacturing Company, Milwaukee, Wisconsin, under NASA Contract NAS8-5392. The work was administered under the direction of the Electrical Components and Power Supplies Section, Astrionics Division, NASA, Huntsville, Alabama. Mr. Eugene Cagle is the technical supervisor for NASA.

This First Quarterly Report covers the work completed from 1 May 1963 to 31 July 1963.

Management direction at Allis-Chalmers includes Mr. W. Mitchell, Jr., Director of Research; Dr. P. A. Joyner, Mr. D. T. Scag and Mr. W. W. Edens, Assistant Directors of Research. The project is supervised by Mr. J. L. Platner, Section Head; and Mr. P. D. Hess, Chief Engineer. The Project Leader is Mr. D. P. Ghere.

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#### ABSTRACT

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A "Static Moisture Removal System" for the capillary hydrogenoxygen fuel cell is described. Feasibility tests and breadboard tests of this system are discussed.

In this system, a desired water vapor pressure is maintained in the cell through a water transport membrane associated with each cell. Water will not be evaporated from the cell electrolyte until the electrolyte vapor pressure exceeds the desired value. Above this value, evaporation rapidly increases.

AUTHOR

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# 1.0 SUMMARY

The theory of operation of the Allis-Chalmers hydrogen-oxygen capillary membrane fuel cell with Static Vapor Pressure Control to accomplish product water removal is presented.

To test the feasibility of the Static Vapor Pressure Control System, three single cells and six four-cell modules were operated. All the cells performed satisfactorily for the duration of the tests which varied in length from 60 to 220 hours. Peak current densities ran as high as 214 ma/cm<sup>2</sup>.

One four-cell and two eight-cell modules were operated in a breadboard system. The first eight-cell module operated under a cyclic load for a total of 405 hours.

A gas chromatograph is being used to experimentally verify the theory of operation of the Static Vapor Pressure Control System.

## 2.0 INTRODUCTION

This is the first quarterly report submitted under Contract Number NAS8-5392. The purpose of the contract is to perform research and development to determine the optimum design approach for Energy Conversion Systems for Advanced Saturn Vehicle Missions.

To accomplish this purpose, studies or experimental investigations will be made in the following general areas:

- (a) A weight and feasibility study will be conducted on a system combining a fuel cell and a cryogenic hydrogen-oxygen heat transfer cycle. The Lunar Logistics Mission concept will be used as a typical mission on which to base the study.
- (b) Experimental and theoretical investigations will be performed on a hydrogen-oxygen capillary fuel cell employing Static Vapor Pressure Control for product water removal. The feasibility and performance characteristics of this system will be investigated for possible application as a vehicle auxiliary power source.
- (c) Comparison studies will be conducted on the Static Vapor Pressure Control fuel cell system with the recirculating reactant Vapor Pressure Control fuel cell under development for MSFC. The study shall show the advantages and disadvantages of each system considered as a complete vehicle subsystem. Mission limitations relative to the Lunar Logistics Vehicle concept shall be given for each system.
- (d) A study will be made of other possible energy conversion systems which may compare favorably or may be used advantageously with the fuel cell system.

The comparison studies will be general in nature and will pertain to weights, volumes, loads, duration of operation, and operational difficulties. These studies will be performed after initial feasibility studies and experimental investigations have been made for the fuel cell system employing Static Vapor Pressure Control.

This report contains the work accomplished under Item (b) above, and includes the theory of Static Vapor Pressure Control Concept, the investigations being conducted, and a summary of feasibility and breadboard testing accomplished during the quarter.

# 3.0 STATIC VAPOR PRESSURE MOISTURE REMOVAL CONCEPT

A fuel cell is an electrochemical converter that converts the chemical energy of fuels directly into electrocal energy, reaction products and some excess heat. In the case of the hydrogen-oxygen fuel cell the reaction product is water.

Efficient and sustained operation of the hydrogen-oxygen fuel cell requires that the product water and the heat produced be removed from the fuel cell system in a controlled manner.

The Static Vapor Pressure Control Concept is a method whereby the product water formed in a capillary fuel cell is removed from the fuel cell stack entirely by diffusion mechanisms and as such is static in nature. The water is removed from the cell stack in the vapor state and may be ejected directly to space, thereby significantly reducing the heat burden imposed upon a fuel cell cooling system. As an alternative the water vapor may be condensed, and in this case the advantages of the static system are applicable, but the thermal advantage is not gained.

The following discussion presents the Static Vapor Pressure Control Concept.

# 3. 1 Capillary Membrane Fuel Cell

The basic cell consists of two porous nickel electrodes separated by the capillary membrane which holds the electrolyte.

The distinctive feature of this cell is the capillary membrane. The capillary potential of the membrane, defined as the differential pressure required to force liquid from its largest pore, is approximately 100 psi. This property enables the membrane to hold the electrolyte in its proper position between the electrodes,

effectively separates the reactant gases and allows communication of water and hydroxyl ions between the electrodes.

When a cell is assembled, the asbestos membrane is filled with electrolyte and swells to a larger volume than when dry. During assembly the associated electrodes are compressed against the asbestos membrane and some of the electrolyte is squeezed from the membrane into the electrodes, forming the necessary catalyst-electrolyte interfaces. The asbestos membrane remains filled with electrolyte.

In an operating hydrogen-oxygen cell, water is formed at the interface of catalyst, electrolyte and hydrogen. If not removed, the water dilutes the electrolyte until it fills the entire electrode pore volume. If the water is removed from the cell faster than it is formed, the electrolyte volume decreases to the point where the wetting front begins to recede into the asbestos. Both overwetting and overdrying are accompanied by rapid deterioration of cell performance.

The capillary membrane fuel cell is capable of satisfactory performance over a range of electrolyte concentrations and volumes. The range of controllable electrolyte concentrations is a function of thickness and porosity of the fuel cell electrodes and the amount of concentration of the electrolyte added to the capillary membrane at the time of construction. Experiments have shown that the average electrolyte concentration should be maintained between 27% and 45% for adequate performance, with a concentration of 35% being optimum. The typical performance of an individual cell as a function of average electrolyte concentration is shown in Figure 1. If the temperature is held constant, then the water vapor pressure of the electrolyte increases rapidly as the electrolyte becomes more dilute, as shown in Figure 2. This characteristic forms the basis for Static Vapor Pressure Control.

# 3.2 Principle of Operation of Static Vapor Pressure Control

The Static Vapor Pressure Control System utilizes the principles of gas and gas-liquid diffusion, the changing vapor pressure characteristics of the Allis-Chalmers fuel cell under load, and the capillary nature of the KOH impregnated membrane.

In this system another asbestos capillary membrane, a water transport membrane, containing a concentrated KOH solution is placed adjacent to, but slightly separated from the hydrogen electrode, as illustrated in Figure 3. Hydrogen and oxygen reactants are dead-ended into the cell. A cavity located behind the water transport membrane is evacuated to a pressure corresponding to the vapor pressure of the desired maximum concentration of KOH solution to be maintained in the cell. A differential pressure of about 23 psi exists across the water transport membrane, but this presents no problem since pressures of over 100 psi are required to expel the KOH solution from the water transport membrane. Hydrogen will not pass through the water transport membrane because the pores of the membrane are complete filled with KOH solution. Water vapor is transported from the cell electrode to the water transport membrane by diffusion through the reactant gas due to the difference in vapor pressure between the cell electrolyte and the KOH contained in the water transport membrane. The water vapor condenses and tends to dilute the KOH in the water transport membrane. This causes the pressure in the evacuated cavity adjacent to the water transport membrane to rise. As the water vapor pressure rises, water vapor is removed from the cavity (and hence from the cell) to an external condenser where it may be condensed to potable water. If there is no use for the water, it may be purged directly to space and by so doing remove approximately 35% of the heat formed within the cell.

In theory, the controlled removal of water from the cell may be accomplished simply by maintaining a fixed pressure upon the water transport membrane corresponding to the vapor pressure of the maximum concentration of the electrolyte desired in the operating cell. The maintenance of the fixed pressure prevents excess water removal. When the cell operates under load and produces water, the subsequent dilution of the cell electrolyte and rapid rise in electrolyte vapor pressure provides the driving force forwater removal.

The transport of water through the water transport membrane and across the cell electrolyte membrane produces KOH gradients in the cell. An illustrative example of the gradient existing in an operating cell is shown in Figure 4.

#### 3. 3 Thermal Considerations

A thermal advantage may be gained with the Static Vapor Pressure Control System as has been stated. Also, because KOH vapor pressure is a function of temperature, the successful operation of this static system requires temperature control with acceptable limits.

The following discussion defines the thermal advantage that may be gained and predicts the allowable temperature variation in an operating cell.

Characteristic of fuel cell operation, some waste heat is produced during the fuel cell reaction. The amount of waste heat produced depends on the thermal efficiency and the power output as shown in the following analysis.

The thermal efficiency, assuming 100% current efficiency, is defined as:

$N_{t}$	<del>,</del> <del>,</del>	Δm	<u>F</u> ΔH°	x	100	ΔH°	=	Enthalpy of reaction per mole of product
						ΔF°	=	Observed free energy change (Observed electrical work)
						∆m	=	Determined moles of reactant consumed

Explicit definition of  $\Delta H^{\circ}$  and  $\Delta F^{\circ}$  requires an explicit definition of reactions taking place in the fuel cell. Operating temperature must also be defined since the thermodynamic properties ( $\Delta F^{\circ}$  and  $\Delta H^{\circ}$ ) are temperature dependent properties. Therefore, for the following discussions the efficiencies are based on the following defined reactions:

$$H_2$$
 (aq.) + 1/2 O<sub>2</sub> (aq.)  $\xrightarrow{25^{\circ} C}$   $H_2O$  (liq.)

Since

 $\Delta F \approx -n \Delta m f E$ ,

where n = equivalents per mole

of reactant

f = Faradays constant

96,500 coulombs per

equivalent

E = observed voltage

 $\Delta F$  observed is proportional to E and  $\Delta F^\circ$  is proportional to E° (1.23 volts). Thus, with reactants and products at 25° C, the thermal efficiency may be determined from the relationship  $N_t = E/1.23 \times \frac{\Delta F^\circ}{\Delta H^\circ}$  or  $N_t = .67 E$ , where E/1.23 is the ratio of the actual cell voltage to the theoretical cell voltage. The last term,  $\frac{\Delta F^\circ}{\Delta H^\circ}$ , is the maximum possible thermal efficiency. The total heat produced is dependent on the load and efficiency as previously stated, and can be easily determined from the relationship:

$$U = P \left(\frac{1}{N_{t}} - 1\right)$$

or

$$U = P \left(\frac{1}{.67 E} - 1\right)$$
 where  $P = power in watts$ 

In a space application, heat energy will be ejected from the system with the water vapor, if the water vapor is vented to space, thereby significantly reducing the heat burden to be dissipated by a vehicle cooling system. The amount of heat energy exhausted in this manner is in direct relationship to the amount of water produced in the cell and to the cell temperature.

The relationship for determining the amount of water produced in an operating cell is:

W = 0.337 A N where A = ampere hours

N = number of cells

W = grams of water

When the total heat energy of vaporization of water at 93.3° C is taken as 0.757 watt-hours/gram, then the heat rejected with the vapor is:

 $U' = 0.256 \times A N$  where U' = watt-hours

The heat rejected in this manner usually amounts to about thirty-five percent of the total heat burden.

If the water vapor is condensed and collected, the heat of condensation must be rejected from the fuel cell system by the fuel cell cooling system and the thermal advantage is no longer gained.

The maximum permissible temperature variation may be predicted by referring to Figure 2. Assuming an acceptable operating range of KOH from 32 to 40 percent KOH and operation in the temperature range of 90° C the permissible temperature variation is 5° C about an optimum KOH concentration of 35 percent.

### 4.0 FEASIBILITY TESTS OF STATIC VAPOR PRESSURE MOISTURE REMOVAL

Feasibility testing of the Static Vapor Pressure Moisture Removal System was conducted to accomplish the following:

- (1) Verify the diffusion transport of the product water from the cell.
- (2) Determine if the rate of water removal by static means was rapid for practical current densities.
- (3) Evaluate water removal from both the hydrogen and oxygen electrode.
- (4) Evaluate the control of multi-cells compared to single cells.

The testing was conducted in two phases. The first phase utilized single-cell units. Although in some cases cell construction differed between tests, the same basic control system shown in Figure 5 was used for all tests. In the second phase, four-cell modules were assembled and operated. A brief summary of the tests conducted follows:

#### Test Number 1

A single cell was constructed as shown in Figure 6. The cell operated for a total of 68 hours at a current density of 93.5 ma/cm<sup>2</sup> with an average voltage of .82 volts. Water was removed from the hydrogen side of the cell for 41 hours and the oxygen side for 27 hours.

#### Test Number 2

This single cell, with the same construction as the first cell, operated for a total of 77 hours at a current density of 88 ma/cm<sup>2</sup> with an average voltage of .80 volts. Water was removed from both sides of the cell for the first 34.5 hours and from the oxygen side for the remainder of the time.

#### Test Number 3

A third cell constructed in the same manner as the previous two cells was operated for 135 hours at a current density of 88 ma/cm<sup>2</sup> with an average of 0.81 volts with water removal on the oxygen side except for short periods at the beginning and end of the test.

#### Test Number 4

A four-cell module was assembled as shown in Figure 7. The module operated satisfactorily for approximately 115 hours under an increasing load as shown in Figure 8. The capability of the Static Moisture Removal System to control a multi-cell module under a variety of load conditions was demonstrated in this experiment.

#### Test Number 5

This four cell module was the same as the previous one except that a new technique of sealing the asbestos was used. Comparison of this with the first three tests indicated that separate water removal manifolding for the hydrogen and oxygen water removal cavities may be more desirable than the common manifolding used in the fourth and fifth tests. This would allow control of the respective cavity pressure settings, which may be desirable because of the electrolyte concentration gradient existing across an operating fuel cell.

#### Test Number 6

This four-cell module was operated for 220 hours with a cyclic load profile. Manifolding was altered to permit individual regulation of hydrogen and oxygen cavity pressures. Figure 9 illustrates the overall module performance with the cavity vapor pressure expressed in terms of equivalent KOH concentration. A summary of total module operating time at the various loads is shown in the following table:

Table I Performance Summary of Feasibility Test Number 6

Load Amperes	Voltage (Volts)	Current Density (ma/cm <sup>2</sup> )	Power (Watts)	Time (Hours)
20	3.51	47.3	70.3	77.0
30	3. 38	70.9	101.4	74.5
40	3. 28	94.5	131.0	68.5
Total Time				220.0 hours

#### Test Number 7

This four-cell module was assembled using improved electrodes and a new type plate designed specifically for the Static Vapor Pressure Control System. The new design provided improved heat removal, water removal

and sealing characteristics. The module was operated for over 100 hours at various loads ranging from 10 amps (24 ma/cm<sup>2</sup>) to 90.5 amps (214 ma/cm<sup>2</sup>). Voltage at the 10 amps was 3.70, at 90.5 amps it averaged 3.00, varying between 2.97 and 3.08. Module performance is shown graphically in Figure 10. The performance of this module was definitely superior to previous modules.

#### Tests Number 8 and 9

These two four-cell modules, which were similar to the previous four-cell module, were constructed primarily to try minor changes in construction and assembly procedures. The modules operated for about 60 hours each at various levels of load. There was evidence that a single hydrogen cavity pressure setting may give adequate performance over a wide range of loads.

The feasibility tests verified the theory of Static Vapor Pressure Control. In addition, the diffusion mechanism of water transport was shown to be rapid and capable of supporting current densities in excess of 214 ma/cm<sup>2</sup>. Water removal from either the hydrogen or oxygen electrode is possible. The stability of multicell stacks was good.

#### 5.0 THEORETICAL INVESTIGATIONS

# 5. 1 Gas Chromatograph Studies

A theoretical and experimental program has been initiated to study the operation of single cells using Static Vapor Pressure Control. This method of moisture control relies on complex processes involving heat and mass transfer which are affected by cell temperature distribution.

Development of a functional mathematical model of a cell operating with Static Vapor Pressure Control would allow evaluation of design changes without expensive, time-consuming testing of each modification. Development of such a model requires the experimental evaluation of certain physical quantities. For example, the diffusion coefficient of water in an aqueous KOH solution held in an asbestos membrane must be determined.

These fundamental studies are being performed with the aid of a chromatograph. This instrument enables the measurement of water vapor pressure existing within the hydrogen and oxygen cavities. Knowledge of the vapor pressures, temperatures, and water removal rates will allow calculation of the desired physical quantities.

The techniques developed for studying the Dynamic Vapor Pressure Control method (conducted under Contract Number NAS8-2696) have been adapted to the present effort.

The test apparatus is shown schematically in Figure 11. The apparatus consists of four general sections:

- (1) The fuel cell with necessary controls and instrumentation.
- (2) A temperature-controlled sampling system.
- (3) The gas chromatograph with controls and carrier gas conditioner.
- (4) A calibration bath.

Gas samples are taken from both the hydrogen and oxygen cavities and are analyzed with the gas chromatograph. The water removal cavity pressure is monitored by a pressure transducer. Operation of the cell is thus monitored while load, cavity pressure and cell temperature are varied in turn. The data obtained is reduced and plotted by a computer.

# 6.0 BREADBOARD SYSTEM TESTING

A control panel has been constructed which forms a complete breadboard system when coupled with a Static Vapor Pressure Controlled fuel cell module. This breadboard, pictured in Figure 12, contains all the components necessary for automatic operation and requires only hydrogen, oxygen and vacuum inputs. However, for laboratory use, heaters and timers are supplied from standard 110 V a.c. instead of being powered by the module. Many instruments are also included for laboratory purposes. (A schematic of the system is shown in Figure 5)

A breadboard system comprised of this control panel and an eight-cell module capable of an output of 400 watts was operated continuously under load except for weekent shutdowns for a total of 405 hours.

The cells which comprised this module contained the same active area (420 cm<sup>2</sup>) as the four-cell feasibility test modules. However, the eight-cell module was designed for moisture removal from the hydrogen electrodes only, hence no water removal cavity existed behind the oxygen plates. The member which contains the oxygen electrode of one cell also contained the water removal cavity for the adjacent cell.

This was the first multi-cell module to be operated with moisture removal from the hydrogen electrode only. It was therefore necessary to show that sufficient moisture could be removed in this manner at all load levels.

It was also desired to obtain data concerning: Optimum and limiting values of cavity pressure; required reactant purge rates; and temperature distribution within the module end cells.

In the first part of the test (162.5 hours) the module was operated on a load program in which the load was increased in 10 amp increments from 20 to 70 amps.

each load being maintained for approximately 24 hours. During the run at each load, the cavity pressure was varied in an effort to seek optimum cell performance.

During the next 122.5 hours of operation, an attempt was made to duplicate the performance of the first hours while using a constant cavity pressure equivalent to an average electrolyte concentration of 43% KOH at the cell operating temperature of 91° C, the value which appeared to be optimum for 70 amp operation. The module was again operated for 24-hour periods at 20, 30, 40, 50, and 60 amps.

During the final 120 hours of the test, the module was operated continuously at 40 amps (95.5 ma/cm<sup>2</sup>). The combined time of all the tests plus miscellaneous operations totalled 405 hours under load. Module performance in terms of voltage, current and cavity vapor pressure (% KOH) is shown in Figure 13, for the first 162.5 hours of operation. This period of operation is typical for the 405 hours.

The above test was significant for two reasons:

- (1) This represented the longest period of operation of fuel cell modules controlled by the Static Vapor Pressure Method.
- (2) The feasibility of operation a multi-cell unit with moisture removal from the hydrogen electrode only was demonstrated.

A second eight-cell module was assembled and tested in the breadboard setup. This module was the same as the previous one except that new gasket material (Butyl rubber) and a new method of thermocouple installation were used.

The module operated under a random cyclic load for a total of 205 hours.

Voltage variation between cells was the smallest so far experienced for a module of this size. The spread between the highest and lowest cell ranged from 20 millivolts at 30 amps to 60 millivolts at 90 amps.

The new (butyl) gasket material showed very good resistance to corrosion by KOH and oxygen and was in excellent shape at the time of disassembly.

Measurement of temperatures in the center and at the surface of selected cells of the module was successfully performed. Preliminary observations showed temperature gradients to average 3° to 8° C from center to edge of cell and 0° to 4° C between cells. As expected, the temperature gradient from center to edge of cell increased proportionately with load.

Purge data is also being analyzed and will be reported later.

#### 7.0 CONCLUSIONS

The Static Vapor Pressure Moisture Removal System has been proven by feasibility and breadboard tests to be operable over a range of cell loads up to 214 ma/cm<sup>2</sup>. Tests on the breadboard system have further simplified the system by showing that water removal on only one side of the cell with a single cavity pressure setting for loads varying over a broad range is possible.

The Static System is well suited for operation in a space environment since all fluid transport occurs in a single gas state and all liquid in the system are contained in capillary pores. Where recovery of product water is not desired, a significant thermal advantage is realized with this system. About thirty-five percent of the waste heat burden produced in the cell may be ejected directly to space vacuum as latent heat of vaporization of the product water, thus reducing the heat burden imposed upon the space vehicle heat rejection system.

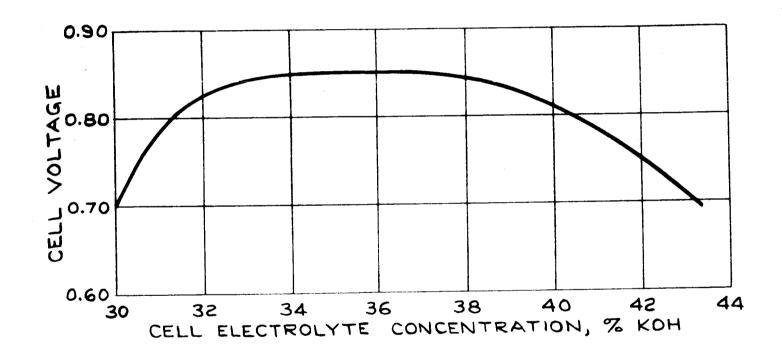
The Static Vapor Pressure Control method eliminates the requirements of recirculating reactant pumps, condensers and gas-water separators. The absence of these components and their associated parasitic power requirements, valves, piping and connections makes the Static Moisture Control System an inherently more efficient and reliable system.

# 8.0 FUTURE WORK

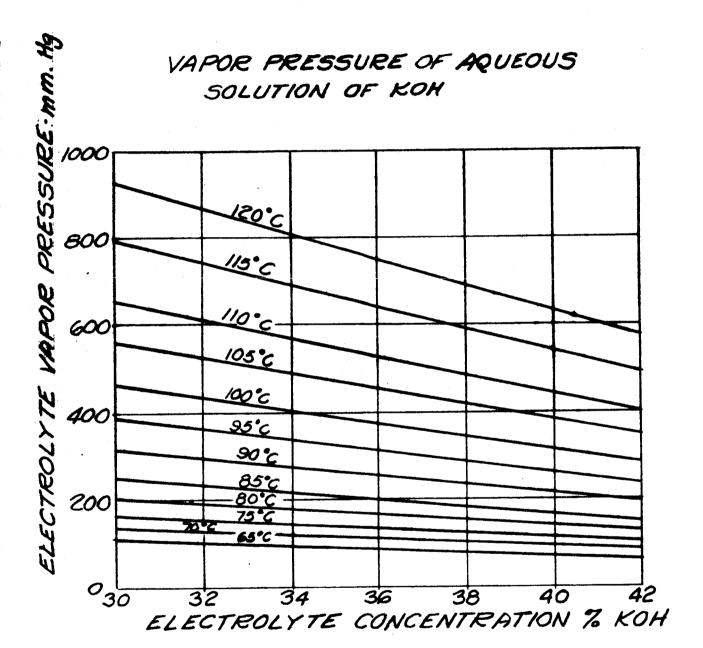
During the next quarter, the following work is planned:

- (1) Continue gas chromatograph studies of Static Vapor Pressure Control.
- (2) Continue study of a combined fuel cell system and cryogenic hydrogen-oxygen heat transfer cycle.
- (3) Continue testing Static Vapor Pressure Controlled cells.
- (4) Start comparison studies.

# FUEL CELL PERFORMANCE AT VARIOUS ELECTROLYTE CONCENTRATIONS



CELL PRESSURE = 30 PSIA CELL TEMPERATURE = 200° F.



# FUEL CELL CONSTRUCTION USING STATIC MOISTURE REMOVAL SYSTEM

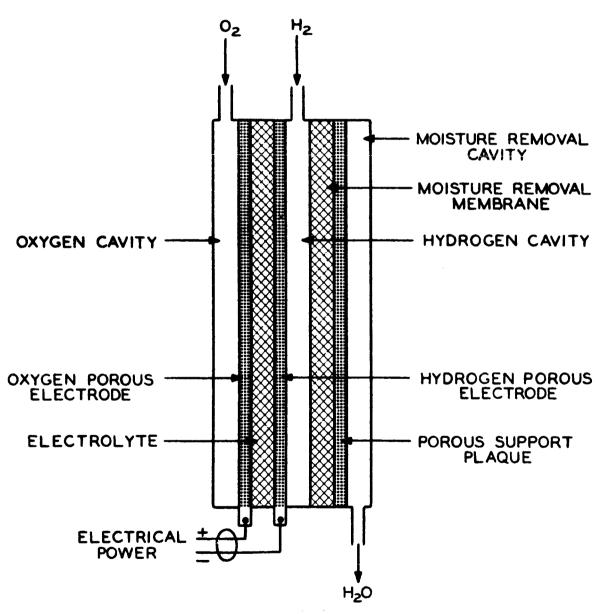


FIGURE 3

# DIAGRAM OF KOH GRADIENTS IN FUEL CELL WITH STATIC VAPOR PRESSURE CONTROL

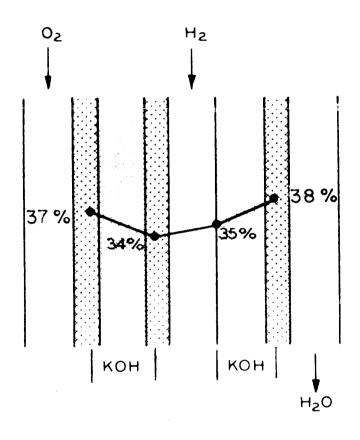


FIG.4

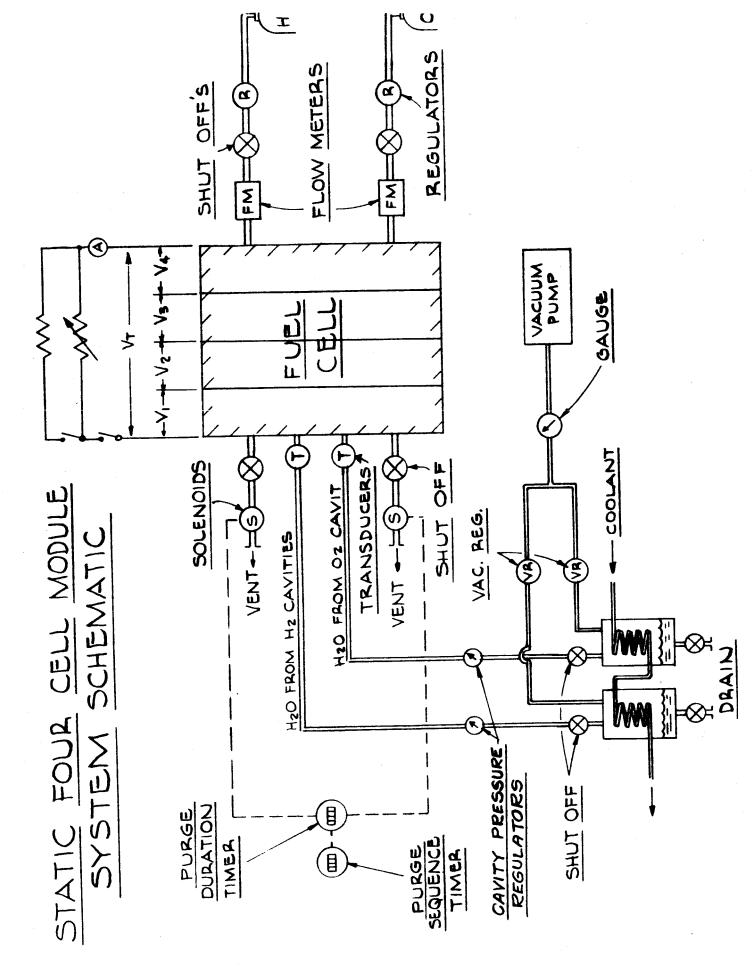
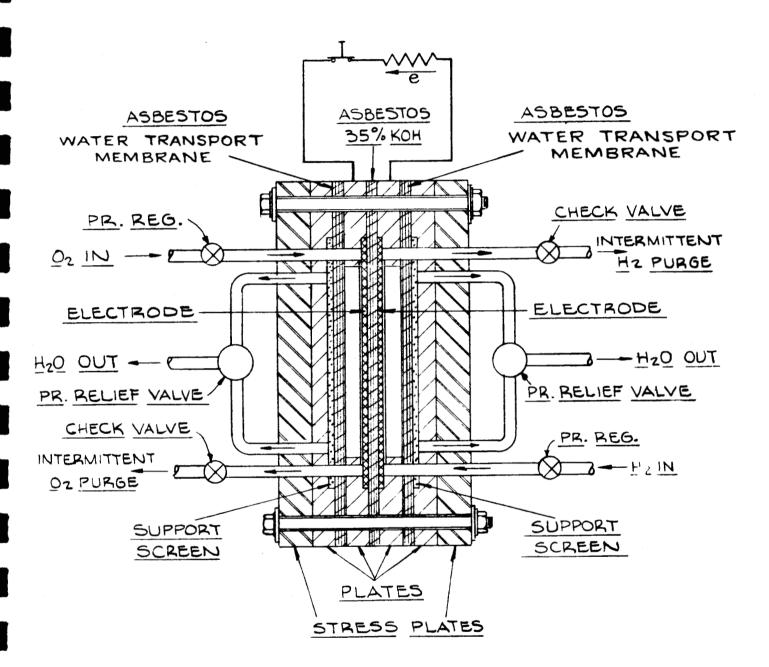
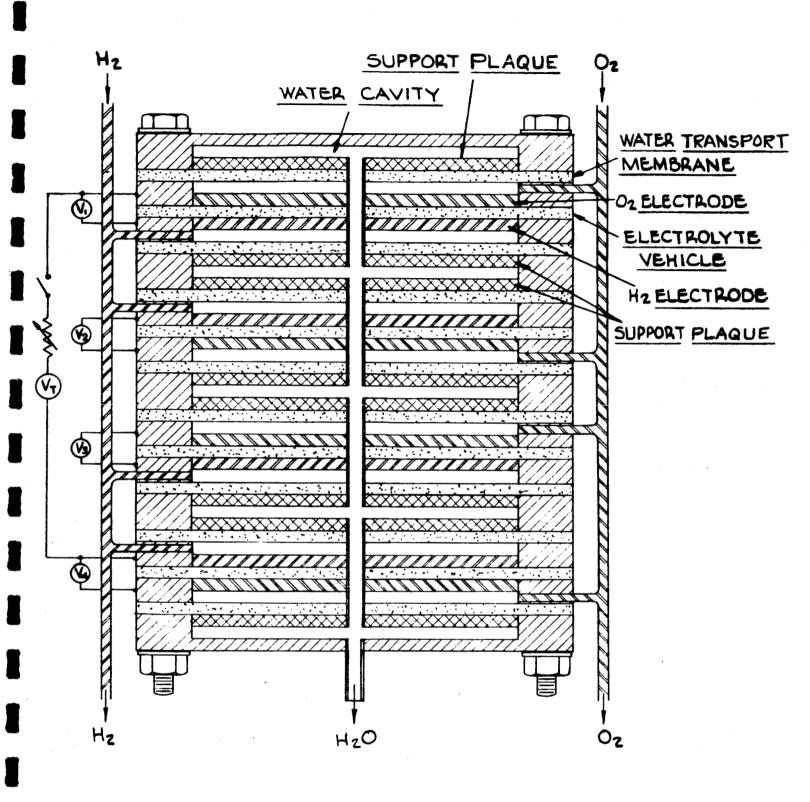


FIG.Nº 5

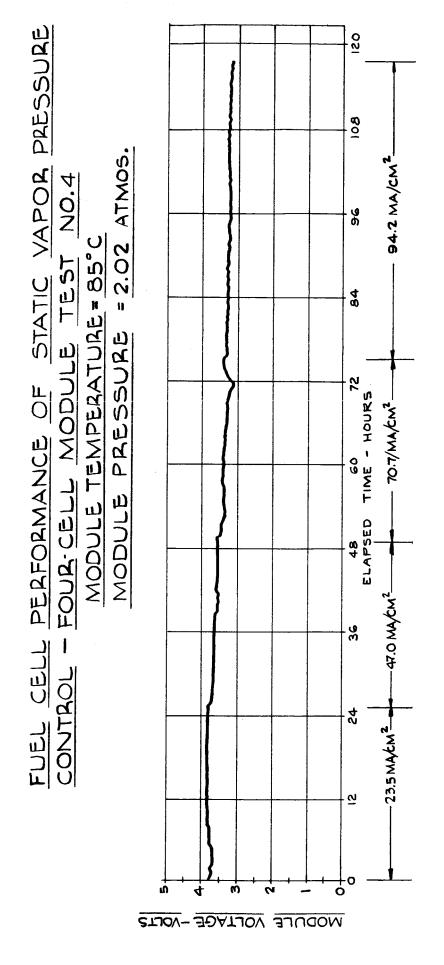
15K63128-6

# STATIC MOISTURE REMOVAL SYSTEM FOR THE H2-02 FUEL CELL

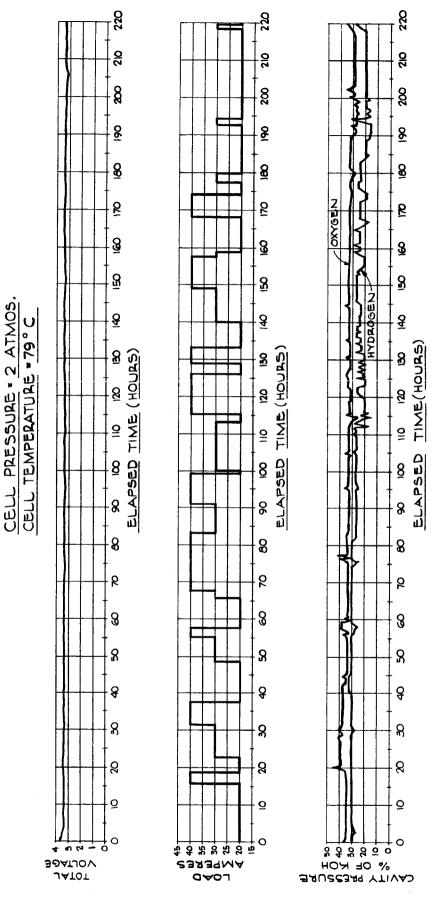




STATIC MOISTURE REMOVAL SYSTEM
FOR THE H2-O2 FUEL CELL
FOUR CELL MODULE



STATIC VAPOR PRESSURE CONTROL



8 8 9.W.H. 510-63 NG58-2696 2.5K63/30-2 **[8**] 8 8 R 3/8 ‡8 g. B 8 g HOURS OF OPERATION \$ 8 8 8 R 9 S 8 g Q 53334W6/71W STOOM TWLOL

FIG. Nº 10

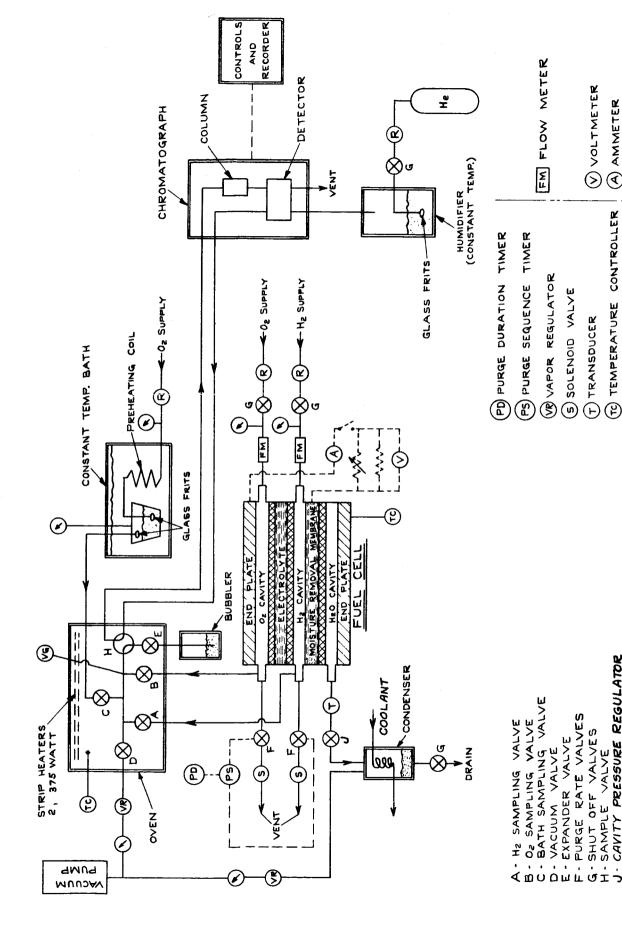
CELL PRESSURE : 2.54 ATM CELL TEMPERATURE - 35.4 °C STATIC MADE PRESSURE CONTROL











VOLTMETER

A AMMETER

CONTROLLER

W VACUUM GAUGE TEMPERATURE

R REGULATOR

T) TRANSDUCER

(E)

CAVITY PRESSURE REGULATOR

VALVE

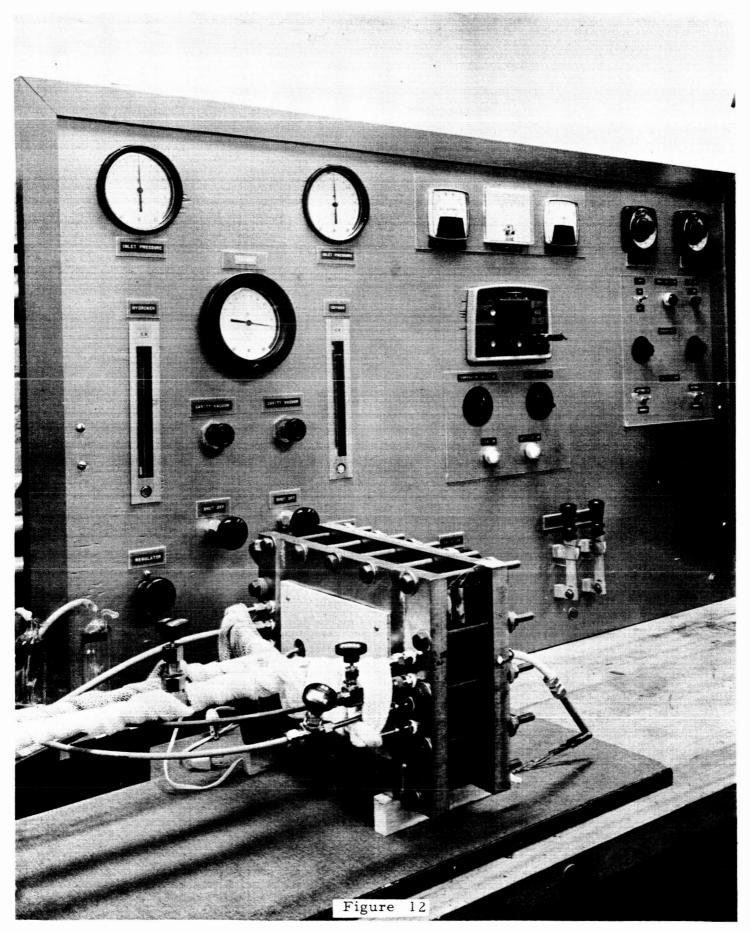
SAMPLE

SHUT OFF VALVES

⊗ NEEDLE VALVE

SAUGE

FIG. Nº 11



STATIC VAPOR PRESSURE CONTROL -- BREADBOARD SYSTEM

